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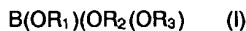
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**CH DE FR GB IT LI**(71) Applicant: **SHIKOKU CHEMICALS  
CORPORATION  
147-1-Minato-machi  
Marugame-shi  
Kagawa-ken (JP)**(72) Inventor: **Nakatsuka, Takuo  
11-121, Nishishin-machi,  
Shiraishi  
Okayama-shi,  
Okayama-ken (JP)  
Inventor: Toyota, Tsuyoshi**

**56-2-203, Saryo, Kinashi-cho  
Takamatsu-shi,  
Kagawa-ken (JP)  
Inventor: Tanimoto, Mie  
2-6-17, Maeshioya-cho  
Marugame-shi,  
Kagawa-ken (JP)  
Inventor: Matsumoto, Akiko  
4689, Fuchu-machi  
Sakalde-shi,  
Kagawa-ken (JP)**

(74) Representative: **Cresswell, Thomas Anthony  
J.A. Kemp & Co.  
14 South Square  
Gray's Inn  
London WC1R 5LX (GB)**(54) **One component epoxy resin compositions containing modified epoxy-amine adducts as curing agents.**

(57) A one component epoxy resin composition comprising, in addition to the epoxy resin, (1) an epoxy adduct obtainable by reacting an epoxy compound having one, two or more epoxy groups per molecule with a nitrogen-containing heterocyclic compound, an aliphatic amine or an aromatic amine, and (2) a boron compound of formula (I)



wherein R<sub>1</sub> to R<sub>3</sub> are the same or different and each is hydrogen, straight-chained, branched or cyclic alkyl or aryl, which composition optionally further comprises (3) a phenolic compound and/or (4) other epoxy curing agents.

The composition is storage stable and cures quickly upon heating.

**EP 0 659 793 A1**

Background of the Invention

## (Field of the Invention)

5 The present invention relates to a one-liquid type epoxy resin composition which can be stored for extended periods of time at ordinary temperature and cures quickly upon heating.

## (Prior Art)

10 Epoxy resins of the two-liquid type have heretofore been used being comprised of an epoxy resin and a curing agent. The epoxy resin composition of the two-liquid type, however, require such operations as weighing, stirring and mixing at a time when they are used on the site. These operations involve such problems as a decreased operation efficiency, erroneous weighing, or disposal of excess amounts of the epoxy resin composition. At present, therefore, study has been extensively conducted in an effort to  
 15 developing one-liquid type epoxy resin compositions that permit highly efficient workability on the site and can be stored for extended periods of time to substitute for the two-liquid type ones.

Development of a latent epoxy curing agent is a prerequisite for preparing the one-liquid type epoxy resin compositions. Known examples of the latent epoxy curing agent include dicyandiamide, carboxylic acid hydrazide, guanamine compounds, imidazole compounds. However, these latent curing agents cure  
 20 the composition by heating at relatively high temperatures for extended periods of time. It has therefore been urged to provide a curing agent that activates at a low temperature and exhibits an excellent latent property.

As latent curing agents for curing epoxy resins at relatively low temperatures, there have been known an adduct of an alkylamine and an epoxy resin, and an adduct of an imidazole compound and an epoxy  
 25 resin (U.S. Patent No. 4,066,625). These adducts only, however, are not enough for obtaining latency to a satisfactory degree. Therefore, additional modification have been reported in which the surfaces thereof are coated with various compounds. For instance, there have been reported a method of encapsulation with an isocyanate compound (Japanese Laid-Open Patent Publication No. 268723/1986) and a method of surface treatment using an acidic substance (Japanese Patent Publication No. 18326/1990, Japanese Laid-Open  
 30 Patent Publication No. 223027/1988).

However, the object has not yet been achieved to a sufficient degree by the above-mentioned attempts of reliably obtaining the latency by the surface treatment. For instance, the capsule film of the isocyanate compound is easily destroyed by the mechanical impact but is not sufficiently regenerated. Besides, the treatment with an acidic compound such as an organic acid, an inorganic acid, a resin acid or an aluminum  
 35 compound does not help obtain stabilized latency.

Summary of the Invention

The object of the present invention therefore is to develop an epoxy resin-curing agent which is free  
 40 from the problems inherent in the prior art by using an epoxy resin adduct (hereinafter referred to as epoxy adduct) that is obtained by adding alkylamines, aromatic amines or nitrogen-containing heterocyclic compounds that are effective as low-temperature curing agents to an epoxy resin, and to develop a one-liquid type epoxy composition that can be stored for extended periods of time by utilizing the above curing agent.

45 Any one of a nitrogen-containing heterocyclic compound, an aliphatic amine or an aromatic amine is reacted with an epoxy compound having one or two or more epoxy groups in one molecule to obtain a product which is then pulverized thereby to afford an epoxy adduct in a powdery form. Surfaces of the powdery epoxy adduct are then coated with a boric acid compound represented by the following general formula to give a latent curing agent.

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$$B(OR_1)(OR_2)(OR_3)$$

wherein  $R_1$  to  $R_3$  are hydrogen atoms, alkyl groups or aryl groups.

There is further proposed an epoxy resin composition using the above curing agent.

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Detailed Description of the Invention

The boric acid compound used in the present invention not only represents boric acid and boric acid ester but also includes the case where boric acid reacts with an alcohol to form a boric acid ester as well as the case where the boric acid ester is, on the other hand, changed into boric acid through hydrolysis. The boric acid compound used in the present invention further includes the case where there exists a so-called partial ester of boric acid. It has in practice been confirmed that when the surfaces of the adduct powder are coated with trimethyl borate, boric acid adheres on the surfaces of the epoxy adduct as confirmed by infrared absorption analysis.

When the surfaces of the very finely granular epoxy adduct are treated with a boric acid ester compound only, the boric acid ester is required in an increased amount, the epoxy-curing property is almost lost. Besides, the surface-treated adduct in an epoxy resin composition aggregates conspicuously upon the curing, causing the resin to be poorly cured. In the acid anhydride-curing system, furthermore, the storage stability is not still satisfactory by a single treatment of boric acid compounds.

In order to solve such problems, the present inventors have attempted to make an epoxy adduct, a boric acid compound, and a phenolic resin present in the epoxy resin composition, and have discovered the fact that the surfaces of the epoxy adduct are coated with the boric acid compound and the phenolic compound, and that the epoxy resin composition can be stably preserved. The inventors have further discovered the fact that even when the epoxy adduct is used as a curing agent or as a cure promoting agent for the acid anhydride-type curing agents, the epoxy resin composition can be stably stored yet exhibiting excellent curing property.

In order to impart latency, furthermore, the boric acid compound and the phenolic compound can be added during the preparation of the epoxy adduct so that they serve as homogeneous components of the epoxy adduct, in spite of coating the surfaces of the epoxy adduct with the boric acid compound and the phenolic compound. However, the reaction product obtained by such a method exhibits epoxy-curing property which is very smaller than that of the surface-coated epoxy adduct.

The powdery epoxy adduct that is used for putting the present invention into practice has an average grain size of from about 0.1 to about 100  $\mu\text{m}$ , and is obtained by pulverizing a product that is obtained by addition-reacting an epoxy compound with alkylamines, aromatic amines or nitrogen-containing heterocyclic compounds according to a conventional method.

The boric acid compound used in Examples of the present invention is represented by the following general formula,



wherein  $\text{R}_1$  to  $\text{R}_3$  are hydrogen atoms, alkyl groups or aryl groups.

Representative examples thereof include boric acid, trimethyl borate, triethyl borate, tripropyl borate, tributyl borate, trioctyl borate, triphenyl borate and tricresyl borate, as well as esters formed by the partial alcohol-exchange reaction.

Representative examples of the nitrogen-containing heterocyclic compound used in Examples of the present invention include imidazole compounds such as imidazole, 2-ethyl-4-methylimidazole, 2-methylimidazole, 2-phenylimidazole, 2-undecylimidazole, 2-heptadecylimidazole, 2-phenyl-4-methyl-5-hydroxymethylimidazole and 2-phenyl-4,5-dihydroxymethylimidazole, as well as mercaptobenzothiazoles, melamine derivatives and the like. It is further allowable to use nitrogen-containing heterocyclic compounds having a 2-hydroxyethyl group on the nitrogen atom such as 1-(2-hydroxyethyl)-2-methylimidazole, 1-(2-hydroxyethyl)-2-phenylimidazole and diethanolamine, since they exhibit actions similar to those of the adduct.

Moreover, representative examples of the aliphatic amine compound and aromatic amine compound used in Examples of the present invention include a diethylenetriamine, a triethylenetetramine, a tetraethylenepentamine, an iminobispropylamine, a bis(hexamethylene)triamine, a diaminodiphenylmethane, a metaphenylenediamine and a 3,9-bis(3-aminopropyl)-2,4,8,10-tetroxaspiro[5.5] undecane.

Representative examples of the phenolic compound used for the epoxy resin composition of the present invention include compounds such as catechol, a 4-*t*-butyl catechol, pyrogallol, resorcin, hydroquinone, phloroglucinol, bisphenol A, bisphenol F, dihydroxybiphenyl, dihydroxynaphthalene, a 1,1,1-tris(4-hydroxyphenyl) ethan and a bis(4-hydroxyphenyl) sulfone, a novolac-type or a resorcinol-type phenol resin, and phenol-type polymers such as a poly(vinyl phenol).

Examples of the epoxy resin used for preparing the epoxy adduct or is blended to the epoxy resin composition of the present invention include a bisphenol A-type epoxy resin, a bisphenol F-type resin, a

cyclic aliphatic epoxy resin, a glycidyl ester-type epoxy resin, a glycidylamine-type epoxy resin, a hydantoin-type epoxy resin, a triglycidylisocyanurate, a phenylglycidylether and a butylglycidylether which may be used in one kind or in a combination of two or more kinds.

In putting the present invention into practice, described below are methods of preparing an epoxy resin composition using an epoxy adduct, a boric acid compound and a phenolic compound as essential components. The methods include (A) a method in which the epoxy adduct, the boric acid compound and the phenolic compound are separately added to the epoxy resin, and are kneaded using a planetary mixer or a roll mill, (B) a method in which a mixture product of the boric acid compound and the phenolic compound is prepared in advance, and the thus prepared product and the epoxy adduct are kneaded together with the epoxy resin, (C) a method in which the surfaces of the epoxy adduct are treated with the boric acid compound only, which is then kneaded together with the phenolic compound and the epoxy resin, and (D) a method in which the boric acid compound and the phenolic compound are reacted with each other, and the obtained reaction product is applied onto the epoxy adduct using a spray drier or the like, which is then kneaded together with the epoxy resin. The surfaces of the epoxy adduct can be coated by using any method of preparing the epoxy resin composition.

When the epoxy resin composition is prepared without adding the phenolic compound, there may be employed (E) a method in which the epoxy adduct and the boric acid compound are separately added to the epoxy resin, and are kneaded by using a planetary mixer or a roll mill, or (F) a method in which the surfaces of the epoxy adduct are treated with the boric acid compound only, and is then kneaded together with the epoxy resin.

In preparing the epoxy resin composition of the present invention, the boric acid or the boric acid ester compound should be blended in an amount of 0.01 to 200 parts by weight and, preferably, in an amount of 0.5 to 40 parts by weight per 100 parts by weight of the epoxy adduct. When the amount of addition is smaller than 0.01 parts by weight, the latency cannot be maintained to a sufficient degree and when the amount of addition is greater than 200 parts by weight, on the other hand, the curing property becomes poor.

The surface-treated epoxy adduct should be blended in an amount of 0.5 to 50 parts by weight and, preferably, in an amount of 1 to 25 parts by weight per 100 parts by weight of the epoxy resin. In this case, suitable curing rate and latent curing property are maintained.

The phenolic compound is blended in an amount of 0.001 to 50 parts by weight and, preferably, in an amount of 0.05 to 10 parts by weight per 100 parts by weight of the epoxy resin. The optimum amount of addition varies depending upon the amount of boric acid or boric acid ester compound that is used at the same time. It is desired that the equivalent ratio of phenolic hydroxyl group to boric acid or boric acid ester is 1:0.2 to 1:5.

According to the present invention, furthermore, an epoxy resin composition that can be stably preserved is obtained even in a curing system in which other epoxy curing agents such as acid anhydrides are added to the above-mentioned epoxy resin composition. Representative examples of the curing agent used here includes acid anhydrides such as maleic anhydride, phthalic anhydride, trimellitic anhydride, pyromellitic anhydride, succinic anhydride, methyl nadic anhydride and chlorendic anhydride, as well as dicyandiamide, acid hydrazides, diaminomaleonitrile, diallylmelamine and amineimides.

In preparing the epoxy resin composition, it is allowable to use, as required, an antioxidizing agent, granular and fibrous fillers of various kinds, and additives such as a plasticizer, a reactive diluent, a coloring agent, a viscosity-adjusting agent and like agents, in addition to the curing agent or the cure accelerating agent.

Contents of the present invention will now be concretely described by way of Examples. It needs not be pointed out that in the present invention, the epoxy resins, amine compounds, nitrogen-containing heterocyclic compounds, phenolic compounds and silica used as starting materials are in no way limited to those described in Examples and Comparative Examples only.

In Examples of the present invention, furthermore, the epoxy adducts are prepared in compliance with a widely known method, and the components constituting the epoxy adducts that are used are described in parenthesis.

#### (1) Epoxy resin

Trade name:	Epikote No. 828, produced by Yuka Shell Co.
Trade name:	Epikote No. 152, produced by Yuka Shell Co.
Trad name:	DER-438, produced by Dow Chemical Japan Co.
Trad name:	DER-331J, produced by Dow Chemical Japan Co.
Trade name:	ERL-4221, produced by Union Carbide Japan Co.

(2) Nitrogen-containing heterocyclic compound

Trade name: Curezol 2E4MZ, produced by Shikoku Kasei Kogyo Co. (2-ethyl-4-methylimidazole)

Trade name: Curezol 2PZ, produced by Shikoku Kasei Kogyo Co. (2-phenylimidazole)

Trade name: Curezol C17Z, produced by Shikoku Kasei Kogyo Co. (2-heptadecylimidazole)

5 Trade name: Curezol C11Z, produced by Shikoku Kasei Kogyo Co. (2-undecylimidazole)

(3) Phenolic compound

Trade name: Resitop PSM-4261, produced by Gun-ei Kagaku Kogyo Co.

Trade name: Resin X, produced by Mitsubishi Yuka Co.

Trade name: Resin M, produced by Maruzen Sekiyu Kagaku Co.

10 (4) Silica

Trade name: Aerosil No. 380PE, produced by Japan Aerosil Co.

The surfaces were treated in compliance with the above-mentioned methods (A) to (D). The storage stability of the epoxy resin composition was evaluated in terms of a gelation time at 40 °C and the curing property of the epoxy resin was evaluated in terms of a relation time at 150 °C. The gelation time was found  
15 by a stroke-cure method on a heated steel plate maintained at a temperature of 40 °C or 150 °C.

(Example 1)

About 280 g of Curezol 2MZ was dissolved in 5 liters of methyl ethyl ketone with heating followed by  
20 the addition of a solution obtained by diluting about 690 g of epoxy resin DER-331J in 500 ml of methyl ethyl ketone. The mixture was refluxed and reacted for two hours. Then, the solvent was removed and the reaction product was finely pulverized to prepare about 950 g of an epoxy adduct.

On the other hand, 2.5 g of Resitop which is a phenolic compound used as a surface-treating agent was dissolved in 25 g of an epoxy resin. 475 Grams of the epoxy resin, 100 g of the above epoxy adduct, 4.5 g  
25 of tributyl borate, and 27.5 g of the above-mentioned epoxy resin in which has been dissolved the Resitop were weighed into a stainless steel pot and were mixed. The mixture was then kneaded at room temperature for about 15 minutes using a three-roll mill to prepare an epoxy resin composition for evaluation.

The gelation times of the obtained epoxy resin composition were measured to be 30 days or longer at  
30 40 °C and 23 seconds at 150 °C.

(Examples 2 to 7)

By using the epoxy adducts shown in Table 1 obtained by the same method as that of Example 1 and  
35 by using the surface-treating agents shown in Table 1, the epoxy resins, epoxy adducts and surface-treating agents were treated at blending ratios shown in Table 1 in the same manner as in Example 1 to prepare epoxy resin compositions. The gelation times of the epoxy resin compositions were measured to be as shown in Table 1.

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Table 1

Example	1	2	3	4	5	6	7
Epoxy resin (Epikote = 828)	100	100	100	100	100	100	100
Epoxy adduct							
(Epikote = 828/2MZ) (Epikote = 828/2MZ/bisphenol A) (Epikote = 828/2PZ) (Epikote = 828/2E4MZ) (Epikote = 828/C11Z) (DER-438/2E4MZ) (ERL-4221/2E4MZ)	20	20	20	20	20	20	20
Surface-treating agent							
Trimethyl borate	0.50	0.50			0.50	0.50	
Tributyl borate	0.90			0.90			
Triphenyl borate					1.10		
Resitop	0.50	0.50	0.50	0.50	0.50	0.50	0.50
Surface-treating method	A	A	A	A	A	A	A
Gelation time							
(sec/150 ° C)	23	72	62	25	95	23	23
(days/40 ° C)	>30	>30	>30	>30	25	>30	>30

(Examples 8 and 9)

By using the epoxy adduct shown in Table 2 obtained by the same method as that of Example 1 and by using the surface-treating agents shown in Table 2, the epoxy resin, epoxy adduct and surface-treating agents were treated at blending ratios shown in Table 2 in the same manner as in Example 1 to prepare epoxy resin compositions. The gelation times of the epoxy resin compositions were measured to be as shown in Table 2.

(Example 10)

2.5 Grams of pyrogallol which is a phenolic compound serving as the surface-treating agent and 2.5 g of a trimethyl borate were dissolved in 40 ml of acetone with heating. Then, acetone was moderately removed by evaporation to obtain a mixture surface-treating agent. Next, 460 g of the epoxy resin, 100 g of the epoxy adduct (curing agent) and 5 g of the above mixture surface-treating agent were weighed into a stainless steel pot and were mixed in advance. The mixture was then mixed at room temperature for about 15 minutes using a three-roll mixer to prepare a sample for evaluation.

The gelation time of the obtained epoxy resin composition was measured to be 30 days or longer at 40 ° C and 24 seconds at 150 ° C.

(Examples 11 to 14)

By using the epoxy adduct shown in Table 2 obtained by the same method as that of Example 10 and by using the surface-treating agents shown in Table 2, the epoxy resin, epoxy adduct and surface-treating agents were treated at blending ratios shown in Table 2 in the same manner as in Example 10 to prepare epoxy resin compositions. The gelation times of the epoxy resin compositions were measured to be as shown in Table 2.

(Example 15)

A solution obtained by dissolving 10 g of triethyl borate in 50 ml of acetone was spray-dried on 100 g of a fine powdery epoxy adduct (DER 331J/2E4MZ) by using a spray drier to obtain an epoxy adduct of

which the surfac has been treated.

To 500 g of the epoxy resin were then added 110 g of the above surface-treated epoxy adduct and 2.5 g of catechol. The mixture was kneaded at room temperature for 15 minutes using a three-roll mill to obtain a desired composition.

5 The gelation time of the resin was 30 days or longer at 40 ° C and was 24 seconds at 150 ° C.

(Example 16)

10 10 Grams of trimethyl borate and 10 g of Resitop were dissolved in 100 ml of acetone, which was then spray-dried onto 400 g of the adduct to obtain the adduct whose surface has been treated.

Then, 110 g of the surface-treated epoxy adduct was added to 500 g of the epoxy resin, and the mixture was kneaded at room temperature for 15 minutes using a three-roll mixer to obtain a desired composition.

15 The gelation time of the resin was 30 days or longer at 40 ° C and was 26 seconds at 150 ° C.

(Example 17)

By using the epoxy adduct shown in Table 2 obtained by the same method as that of Example 16 and by using the surface-treating agents shown in Table 2, the epoxy resin, epoxy adduct and surface-treating agents were treated at blending ratios shown in Table 2 in the same manner as in Example 16 to prepare epoxy resin compositions. The gelation times of the epoxy resin composition were measured to be as shown in Table 2.

(Example 18)

25 The epoxy resin, epoxy adduct and boric acid were mixed together at a blending ratio shown in Table 2 quite in the same manner as that of Example 1 but without using the phenolic compound, in order to prepare an epoxy resin composition. The gelation times of the thus obtained epoxy resin composition were measured to be as shown in Table 2.

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(Example 19)

35 The epoxy adduct of which the surfaces have been treated with methyl borate and the epoxy resin were mixed together at a blending ratio shown in Table 2 quite in the same manner as that of Example 15 but without using the phenolic compound, in order to prepare an epoxy resin composition. The gelation times of the thus obtained epoxy resin composition were measured to be as shown in Table 2.

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Table 2

Example	8	9	10	11	12	13	14	15	16	17	18	19
Epoxy resin (parts) Epikote #828	100	100	100	100	100	100	100	100	100	100	100	100
Epoxy adduct (parts) DER331J/2E4MZ	20	20	20	20	20	20	20	20	20	20	20	20
Surface-treating agent (parts)												
Trimethyl borate	0.40	0.25	0.50		0.46	0.20	0.50		0.50			0.50
Triethyl borate								0.50		0.40		
Boric acid				0.33							1.0	
Resitop	0.80								0.50			
Resin X				0.34						0.40		
Resin M						0.80						
Triphenolethane		0.75										
Gallic acid							0.50					
Pyrogallol			0.50	0.67								
Catechol								0.50				
Surface-treating method	A	A	B	B	B	B	B	C	D	D	E	F
Gelation time (sec/150°C) (days/40°C)	23 >30	26 >30	24 >30	21 17	25 14	22 >30	23 >30	24 >30	26 >30	23 >30	24 1	71 >30

(Examples 20 to 22)

A variety of surface-treated epoxy adducts were obtained in the same manner as in Example 15.

Then, the epoxy resin, phenolic compounds, Aerosil and the epoxy adducts serving as the curing promoting agents were kneaded at blending ratios shown in Table 3 at room temperature for 15 minutes using a three-roll mixer. The kneaded product was then transferred to a planetary mixer followed by the addition of a predetermined amount of methylnadic anhydride which is a curing agent. The mixture was



mixed with stirring under a reduced pressure for 30 minutes to obtain epoxy resin compositions. The gelation times of these epoxy resin compositions were measured to be as shown in Table 3.

(Examples 23 and 24)

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A variety of surface-treated epoxy adducts were obtained in the same manner as in Example 15.

Then, the epoxy resin, phenolic compounds, Aerosil, dicyandiamide as a curing agent and the epoxy adducts as a cure accelerating agents were kneaded at blending ratios shown in Table 3 at room temperature for 15 minutes using a three-roll mixer in order to prepare epoxy resin compositions. The gelation times of these epoxy resin compositions were measured to be as shown in Table 3.

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(Examples 25 and 26)

By using the epoxy adduct shown in Table 3 obtained by the same method as that of Example 1 and by using the surface-treating agents, the epoxy resin, curing agent, epoxy adduct and surface-treating agents were treated at blending ratios shown in Table 3 in the same manner as in Example 1 to prepare epoxy resin compositions. The gelation times of the epoxy resin compositions were measured to be as shown in Table 3.

20

Table 3

	Example	20	21	22	23	24	25	26
	Epoxy resin (parts)							
25	Epikote #828 DER-331J	100	100	100	100	100	100	100
	Curing agent (parts)							
30	Methylnadic anhydride Dicyandiamide	85	85	85	4	4	85	85
	Cure promoting agent (parts)							
35	(DER-438/2E4MZ) (Epikote#152/2E4MZ) (DER-438/2PZ)	6	6	6	5	5	6	6
	Surface-treating agent (parts)							
40	Trimethyl borate Pyrogallol Catechol Resitop Aerosil (parts) Surface-treating method	4 5  3 C	5 5  3 C	5 4 3 C	5 5 2 C	5  2 2 C	4  3 A	5  3 A
45	Gelation time							
	(sec/150 ° C) (days/40 ° C)	330 >30	320 >30	250 >30	96 >30	82 >30	305 5	320 2

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(Comparative Examples 1 and 2)

An epoxy adduct of a composition shown in Table 4 serving as a curing agent or an imidazole compound was mixed at a blending ratio as shown in Table 4 to prepare an epoxy resin composition. The gelation times of the epoxy resin composition were measured to be as shown in Table 4.

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## (Comparative Example 3)

An epoxy resin composition was prepared quite in the same manner as in Example 1 but without adding the boric acid compound. The gelation times of the epoxy resin composition were measured to be as shown in Table 4.

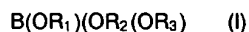
Table 4

	Comparative Example		1	2	3
10	Epoxy resin Epikote #828		100	100	100
	Curing agent				
	Adduct				
15	ERL-4221 (5.1 parts)		15	6.1	20
	C <sub>17</sub> Z (6.1 parts)				
	Pyrogallol (2.0 parts)				
20	Boric acid (1.8 parts)				
	Curezol C <sub>17</sub> Z				
	Adduct (Epikote#828/2E4MZ)				
	Resitop				
25	Gelation time	(sec/150 ° C) (days/40 ° C)	>1800 >30	155 >30	23 1

When an epoxy adduct is used as a curing agent or is used as a cure accelerating agent for other curing agents, boric acid or a boric acid ester compound and a phenolic compound are made present together with the epoxy adduct in the epoxy resin composition, so that the epoxy resin composition exhibits favorable storage stability and curing property. The epoxy resin composition of the present invention, therefore, can be used as an adhesive, a sealant, an ink, a painting material and the like.

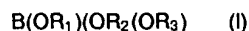
### Claims

1. An epoxy resin composition comprising, in addition to the epoxy resin, (1) an epoxy adduct obtainable by reacting an epoxy compound having one, two or more epoxy groups per molecule with a nitrogen-containing heterocyclic compound, an aliphatic amine or an aromatic amine, and (2) a boron compound of formula (I):



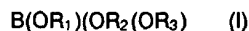
wherein R<sub>1</sub> to R<sub>3</sub> are the same or different and each is hydrogen, straight-chain, branched or cyclic alkyl or aryl, which composition optionally further comprises (3) a phenolic compound.

2. A latent epoxy curing agent or a cure accelerating agent comprising a fine granular epoxy adduct obtainable by reacting an epoxy compound having one, two or more epoxy groups per molecule with a nitrogen-containing heterocyclic compound, an aliphatic amine or aromatic amine, the surfaces of the fine granular epoxy adduct being coated with a boron compound of formula (I)



wherein R<sub>1</sub> to R<sub>3</sub> are the same or different and each is hydrogen, straight-chain, branched or cyclic alkyl or aryl, which composition optionally further comprises (3) a phenolic compound.

3. A surface-treating agent for an epoxy adduct comprising a boron compound of formula (I)



wherein  $R_1$  to  $R_3$  are the same or different and each is hydrogen, straight-chained, branched or cyclic alkyl or aryl, which composition optionally further comprises (3) a phenolic compound.

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4. A process for producing an epoxy resin composition according to claim 1 comprising the step of kneading an epoxy resin with the other components of the composition.

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5. A process according to claim 4 comprising the adding the epoxy adduct, the boron compound and optionally the phenolic compound separately in any order to the epoxy resin.

6. A process according to claim 4 comprising mixing the boron compound and the phenolic compound to form a mixture and adding the mixture and the epoxy adduct separately in any order to the epoxy resin.

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7. A process according to claim 4 comprising surface-treating the epoxy adduct with the boron compound then adding the surface treated-boron compound, and optionally separately adding in any order the phenolic compound, to the epoxy resin.

20

8. A process according to claim 4 comprising reacting the boron compound and the phenolic compound to form a product, surface-treating the epoxy adduct with the product and mixing the surface-treated epoxy adduct to the epoxy resin.

25

9. A process according to any one of claims 4 to 8 comprising reacting an epoxy compound having one, two or more epoxy groups per molecule with a nitrogen-containing heterocyclic compound, an aliphatic amine or an aromatic amine to as to form the epoxy adduct.

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10. Use of an epoxy resin according to claim 1, a latent epoxy curing or cure accelerating agent according to claim 2 or a surface-treating agent according to claim 3 in production of a cured epoxy resin product.

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## EUROPEAN SEARCH REPORT

Application Number  
EP 93 31 0516

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
X	EP-A-0 458 502 (THE DOW CHEMICAL CO.)  * abstract * * page 4, line 25 - page 5, line 41 * * page 11, line 41 - page 14, line 7 * * page 17, line 23 - page 18, line 32 * ---	1-5,7,9,10	C08G59/40 C08G59/56 C08K3/38 C08K5/55
X	PATENT ABSTRACTS OF JAPAN vol. 16, no. 13 (C-0901) 14 January 1992 & JP-A-03 234 727 (THREE BOND CO. LTD.) 18 October 1991 * abstract *	1-4,10	
Y	DATABASE WPI Week 9021, Derwent Publications Ltd., London, GB; AN 90-160746 & JP-A-2 103 224 (TOKYO THREE BOND KK) 16 April 1990 * abstract *	1-3,10	
D,Y	US-A-4 066 625 (J. C. BOLGER) * abstract; claims 1,2 * * column 1, line 18 - column 6, line 68 * * column 10, line 42 - column 11, line 23; examples 2,3 * ---	1-3,10	TECHNICAL FIELDS SEARCHED (Int.Cl.6)  C08G C08K
A	EP-A-0 197 458 (ASAHI DENKA KOGYO K.K.) * abstract * * column 20, line 52 - column 23, line 25 * ---	1,2	
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The present search report has been drawn up for all claims			
Place of search BERLIN		Date of completion of the search 1 June 1994	Examiner Krische, D
<b>CATEGORY OF CITED DOCUMENTS</b>			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		I : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons  * : member of the same patent family, corresponding document	

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Application Number  
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DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
A	DATABASE WPI Week 8835, Derwent Publications Ltd., London, GB; AN 88-245768 & JP-A-63 178 125 (TAOKA CHEMICAL KK) 22 July 1988 * abstract *  -----	2,3,7,8	
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The present search report has been drawn up for all claims			
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